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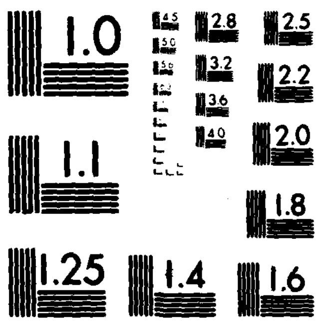

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To appear in Appl. Physics Lett. April '81

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Time-Resolved Optical Transmission of Pulsed Laser-Irradiated Silicon

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15 N00014-80-C-0414

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ABSTRACT

The time-resolved optical transmission of silicon has been observed at λ = 1.15 μm during irradiation by an 8 nsec pulsed laser at 485 nm with several energy densities in the range of .25 to 1.2 J/cm^2. The transmission exhibits a sudden brief drop consistent with the rise and fall of the reflectivity enhancement. However, the transmission does not exhibit the strong absorption expected of molten silicon with a skin depth of ∿ 100Å.

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The experiment most often cited to support the thermal melting model of laser annealing has been the time-resolved reflectivity study of Auston, et al.[1] They showed that the reflectivity rose to a value roughly that of molten silicon for periods of up to ~ 500 nsec. Subsequently, the duration of this enhanced reflectivity has been used to measure the duration of the molten phase by a large number of workers.[2] However, we have demonstrated by Raman scattering measurements[3] that the lattice temperature rise 10 nsec after an 8 nsec "heating" pulse with energy density of ~ 1 J/cm^2 is far below the melting temperature of silicon. The observed lattice temperature rise of only 300°C could be consistent with the existence of a hot, dense electron-hole plasma which exists for tens of nanoseconds and retains most of the pulse energy.[4] The enhanced reflectivity then might be due to a solid-state plasma with densities of the order of several times 10^{21} cm^{-3}. However Nathan, et al.[5] searched for a wavelength dependence in the duration of this plasma reflectivity and found none in contradiction with a simple solid state plasma having a density which is a smoothly varying function of time.

Quite recently[6] we have performed a Raman study of the onset of recrystallization following an 8 nsec annealing pulse incident on ion-implantation-amorphized silicon. We found clear evidence of a large fraction of recrystallized material after 28 nsec and while the reflectivity was still enhanced. (The enhanced reflectivity duration was ~ 50 nsec.) The observation of such phonon-Raman signals during the enhanced reflectivity period raises important questions about the optical skin depth of the Raman probe pulse at 405 nm. For example if the skin depth during this high reflectivity period is much shorter than the normal light penetration depth ($\alpha^{-1} \approx .16$ μm at $\lambda = 405$ nm), the Raman signal should have been greatly attenuated.

Thus, in order to improve our understanding of the Raman results and in order better to understand the origin of the reflectivity enhancement itself we have performed the complementary experiment, viz. to measure the time-resolved transmission of this enhanced-reflectivity phase. Using a probe at $\lambda_1 = 1.15$ μm, we find very little absorption during the enhanced reflectivity phase in contrast with the known skin depth of molten silicon which ranges from 120 Å at 1 μm to 80 Å at 400 nm.[7] This stands in direct contradiction with normal thermal melting models of laser annealing[8] but is consistent with the Raman temperature measurements and the Raman measurement of the onset of recrystallization.

For the transmission measurements we have used essentially the same arrangement as previously used[3,6] for monitoring reflectivity during Raman data acquisition. That is, a helium-neon laser operating at 1.15 μm was focussed to a diameter of 50 μm or less in the center of the pulsed laser spot of diameter ∿ 2 at mm. A nitrogen-laser-pumped dye laser provided the 8 nsec excitation pulse at $\lambda = 485$ nm. Beam overlap and the power density at the center of the pulsed beam were repeatedly checked with a 50 μm diameter pinhole placed exactly at the sample position. A measure of the sizes of the annealing and the probe beams was obtained by the observed ratio of power transmitted through the 50 μm pinhole to the full beam or pulse power. For the CW probe beam the fraction transmitted through the 50 μm pinhole was greater than 75%, whereas for the excitation pulse only 3% was transmitted. An independent check of the pulsed beam spot size and homogeneity was obtained from the size of the annealed or partially annealed spot on a silicon surface amorphized by ion implantation. This slightly elliptical spot measured approximately 300 μm by 400 μm when the power density through the central 50 μm was adjusted to 1 J/cm^2. At an annealing pulse energy density of 1 J/cm^2 in the central 50 μm we found an epitaxial regrowth region of 100-150 μm in diameter indicated by CW Raman polarization measurements. Epitaxial regrowth of the central region was confirmed by TEM measurements.[9] For the surrounding annular region to a diameter of ∿ 400 μm the regrowth appears not to be epitaxial. No evidence of localized hot spots was seen when this beam was gradually attenuated by neutral density filters until even the beam center did not produce any evidence of annealing visible under an optical microscope. The time resolved measurements were performed with the 1.15 μm beam incident at 10° to the normal using a germanium PIN photodiode with a rise time of 3-4 nsec.

The penetration depth of the 485 nm pulsed beam is α^{-1} = 1.1 μm so that if thermal melting occurs one should expect the melt depth at ∿ 1 J/cm^2 to be approximately 1 μm. However if one assumes that only 0.12 μm has melted then the 120 Å skin depth of molten silicon still requires that the transmission at λ = 1 μm would be e^{-10}. Transmission measurements in strongly absorbing media are particularly sensitive to light leakage. Therefore, since we found only very weak absorption at 1.15 μm during the presumably molten phase (vide infra), we repeated the measurements for two different focal spot sizes of the 1.15 μm probe beam. Transmission and reflectivity were measured with the spot diameter increased by $\sqrt{2}$ and decreased by a factor of 2. The results were unchanged.

Transmission and reflectivity of a 400 μm thick unimplanted (10^3 Ω-cm) silicon wafer observed with the 1.15 μm probe are shown in Figure 1. At 1.1 J/cm^2 for the 485 nm pulsed beam, one observes a flat-topped enhanced reflectivity lasting approximately 60 nsec. The transmission shows a sudden drop as the reflectivity rises and a corresponding recovery as the reflectivity falls. It is immediately apparent that the transmission minimum does not in any way approach the e^{-10} expected on the basis of the conservative estimate of melt depth given above. This result stands in direct contradiction to the normal thermal melting hypothesis.

Two additional effects are also evident in Fig. 1. 1) The transmission does not fully recover when the reflectivity does, but instead a residual transmission loss (of the order of 25%) recovers with a time constant of about 400 nsec. (See trace c of Fig. 1). 2) The period of sharply diminished transmission lasting $\sim$ 60 nsec does not possess a horizontal bottom but shows a continuously decreasing transmission. This effect is quite reproducible. It is tempting to separate the transmission behavior into two components. The first factor is due to reflective losses at the silicon-air interfaces for which $T = 1-R$ implies $T_o = 1-.3 = .7$ before the pulse arrives and $T = 1-.5 = .5$ when the high reflectivity phase is reached. The second factor influencing transmission is likely an absorptive component which apparently increases gradually during the highly reflecting phase and decreases thereafter with the observed time constant of $\sim$ 400 nsec. This absorptive component could arise from free carrier absorption due to the carriers generated by the 485 nm pulsed beam. The transmission loss not explained by reflectivity changes corresponds to $\alpha\ell \approx 0.5$ where ℓ is the depth of the absorbing region ($\ell \gtrsim 1\mu m$, the pulsed laser absorption depth). The free carrier absorption due to laser-generated carriers has been measured and yields a cross section per carrier of $\sigma \sim 5 \times 10^{-18}$ cm^2 at $\lambda \sim 1.06\mu$m. (11) Thus the absorptive component seen in Fig. 1 would require a carrier density of $n = \alpha/\sigma \tilde{=} 10^{21}$cm^{-3}. Note that we have not directly measured the depth of the absorbing region and if that depth were somehow less than 1μm the calculated carrier density would be correspondingly increased.

The time-resolved transmission at $\lambda \sim 1.15 \mu m$ was studied for a variety of pulsed laser (485 nm) powers. The features shown in Figure 1 are reproduced except that the reflectivity duration varies smoothly from zero near $0.5 J/cm^2$ to ~ 150 ns near $1.5 J/cm^2$. In all cases the duration of the sharply reduced transmission matches the reflectivity duration. $1.5 J/cm^2$ is the damage threshold as evidenced by permanent surface morphology changes observable by optical microscopy. Above the damage threshold the transmission drops to zero within twenty nanoseconds and <u>never</u> recovers. Thus, we cannot distinguish by transmission measurements a possible strongly absorbing molten phase from surface damage which refracts transmitted light away from the detector.

The principal, striking conclusion of the transmission measurement is that there is no evidence of a molten silicon phase below the damage threshold with absorption in any way approaching that of normal molten silicon with its skin depth of ~ 100 Å. An alternative explanation for the flat-topped enhanced reflectivity signal is the existence of a dense solid state plasma.[4,10] The present data, like the results of Nathan, *et al.*,[5] contradict the results expected from an ordinary Drude model of a simple plasma for the following reason: The fact that we obtained the usual flat-topped reflectivity behavior at $\lambda_1 = 457.9$ nm[12] implies that the plasma frequency exceeds the laser frequency. Thus $\omega_p \geq \omega_L = 4.1 \times 10^{15}$ rad/sec. The Drude model of the electrons then predicts a skin depth which decreases with frequency below ω_p. Thus for a probe at $\lambda_2 = 1.15 \mu m$, one ought to see a skin depth for the light intensity given by

$$\delta = \frac{c}{2(\omega_p^2 - \omega_2^2)^{1/2}} \leq \frac{c}{2(\omega_1^2 - \omega_2^2)^{1/2}} = 400 \text{ Å} \quad . \qquad (1)$$

If the plasma exists to a depth of $\sim \alpha^{-1}$ of the 485 nm excitation pulse, (α^{-1} = 1.1 μm) then severe attenuation of the 1.15 μm probe should occur. This is definitely not observed.

How then are the reflectivity, transmission, and Raman results to be understood? We think that the simultaneous changes in the transmission and reflectivity signatures with their flat, nearly constant profiles in time strongly suggest the occurrence of a transition to a phase with a radically different index of refraction. Van Vechten, et al.,[4,13] in fact, originally suggested the possibility of a new fluid phase arising from a dense, hot plasma. Such a phase transition driven by a dense plasma would produce a flat-topped enhanced reflectivity with a duration not wavelength dependent and would not require the high absorption which is the corollary of a plasma-generated reflectivity. A plasma reflectivity at λ = 457.9 nm requires a density of

$$n = \frac{\omega_p^2 m^* \varepsilon(\infty)}{4\pi e^2} = 5 \times 10^{21}\ \mathrm{cm}^{-3} \qquad , \qquad (2)$$

with $\varepsilon(\infty) = 1$ and $m^* = m_o$. Since the electron system is very dense and presumably very hot the free electron mass, m_o, seems a reasonable choice for m^*. The use of a high frequency dielectric constant of 1 is probably not reasonable but the effect of using the optical dielectric constant ε = 12 raises the estimate of n correspondingly higher. On the other hand Van Vechten and Wautelet[14] have calculated that there will be changes to the electronic energy band structure and a decrease of the band gap sufficient to lead to plasma confinement at significantly lower densities ($\sim 10^{21}/\mathrm{cm}^3$). Plasma densities in the low 10^{21} range would be consistent with the present observations. Dense plasma effects are undoubtedly intimately related to the annealing mechanisms with pulsed lasers and electron beams but much work still remains before we obtain a completely satisfying understanding of the physics involved.

The authors are indebted to J. A. Van Vechten for many stimulating conversations during the course of these experiments. The support of the Office of Naval Research (contract no. N000 14-80-C-0419) is gratefully acknowledged.

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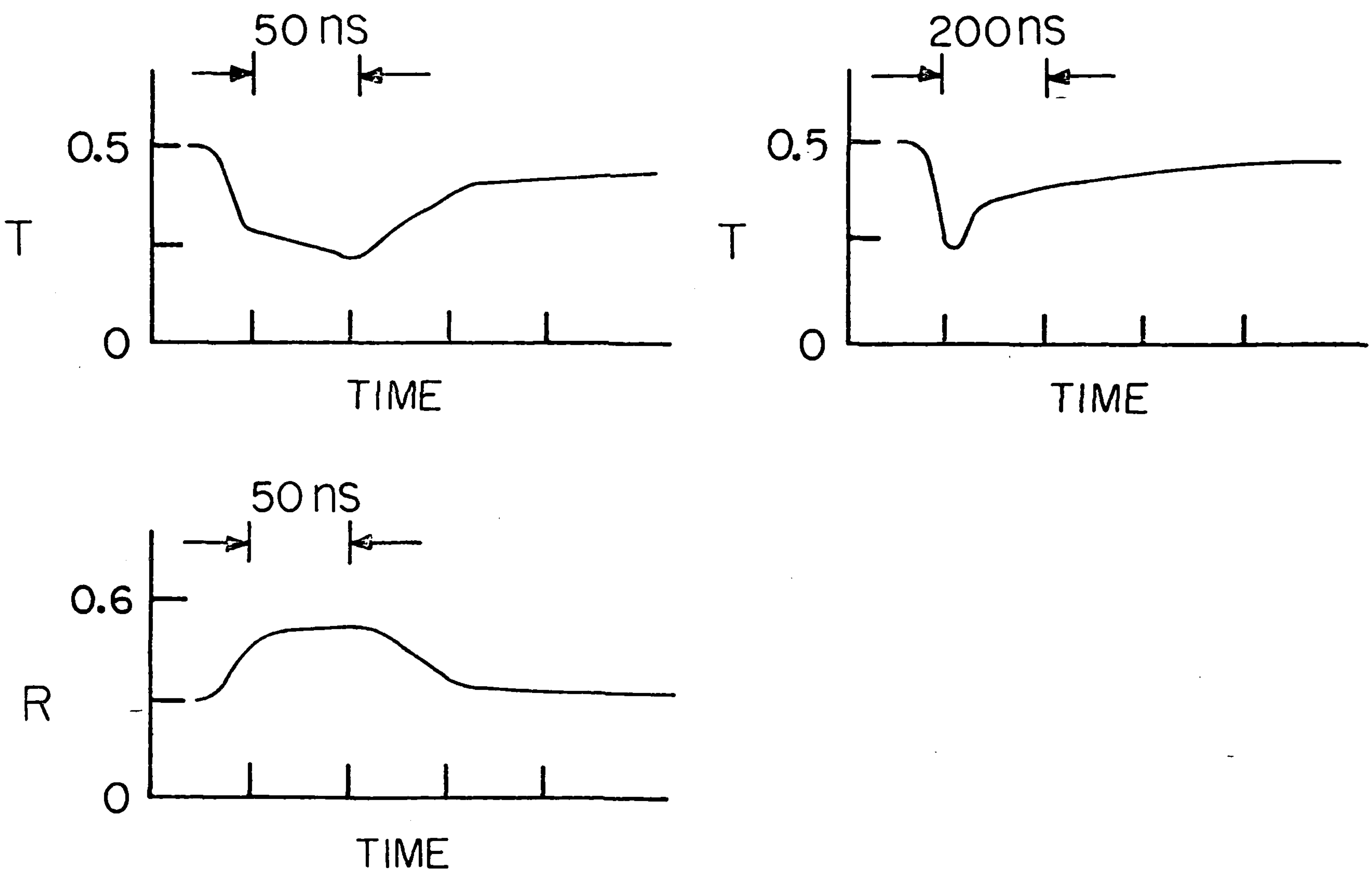
Figure Caption

Fig. 1 Transmission and reflectivity measurements on crystalline silicon (0.4 mm thick) with a CW probe beam of $\lambda = 1.15\ \mu m$ and a 485 nm excitation pulse ($P = 1.1\ J/cm^2$).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